Understanding and Pushing the Limits of Critical Current in Coated Conductors

Steve Foltyn, Leonardo Civale

Focus: Investigating thickness dependence of J_c

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> Superconductivity Technology Center Los Alamos National Laboratory

> > FY 2006 Project Cost: \$500k

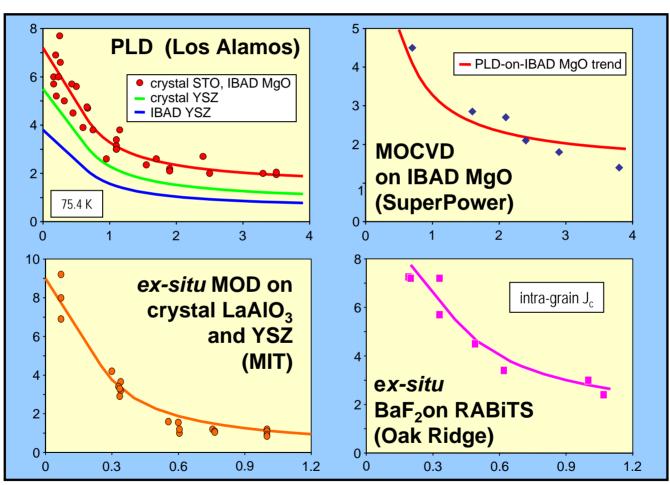
Next talk: Boris Maiorov

"Expanding the set of tools for investigating and improving vortex pinning"



Thickness dependence of J_c can be strikingly similar for a variety of different deposition processes

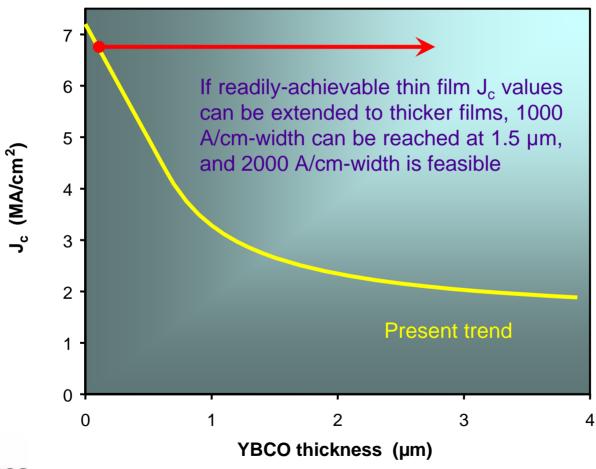
Critical current density (MA/cm²)





YBCO thickness (µm)

The motivation for understanding and controlling thickness dependence is clear





Three models have been proposed to explain thickness dependence

- **★ Intrinsic physics:** J_c is fundamentally related to film thickness through a mechanism such as the Lorentz force growing faster than the pinning force as thickness is increased.
- ★ Microstructural decay: The high quality typical of thin films cannot be maintained in thicker films, so J_c decreases with thickness.
- ★ Interfacial enhancement: Film properties near the substrate differ from those far from the interface in such a way that J_c is higher near the interface.



We will present a summary of existing evidence plus new evidence supporting the third model

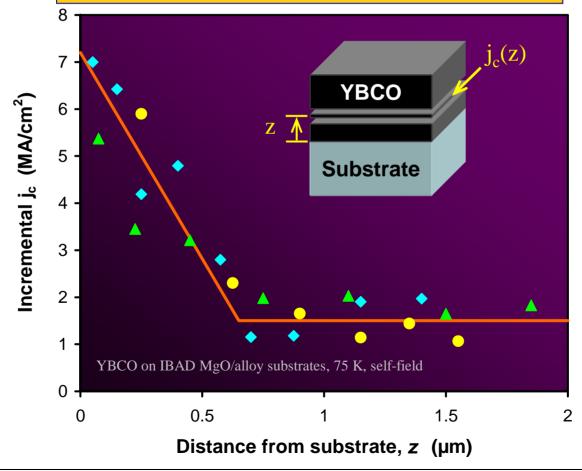
- * Intrinsic physics: J_c is fundamentally related to film thickness through a mechanism such as the Lorentz force growing faster than the pinning force as thickness is increased.
- Microstructural decay: The high quality typical of thin films cannot be maintained in thicker films, so J_c decreases with thickness.
- **★** Interfacial enhancement: Film properties near the substrate differ from those far from the interface in such a way that J_c is higher near the interface.

We first described the interfacial enhancement model at the 2004 Peer Review. Since that time we have developed conclusive evidence that it correctly explains high J_c in thin films and the rapid drop as thickness is increased.



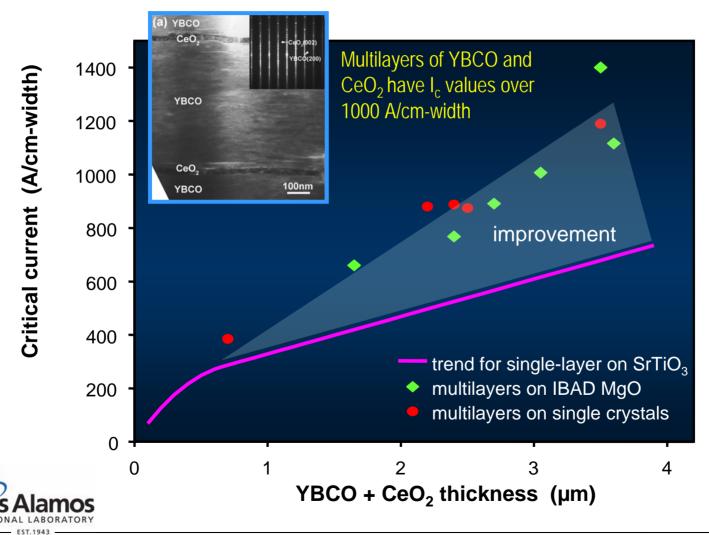
We developed the interfacial enhancement model after evaluating the incremental variation of J_c within a film...

Symbols: Result obtained by ion milling three samples. **Line:** Result obtained by fitting to J_c(t) curve.



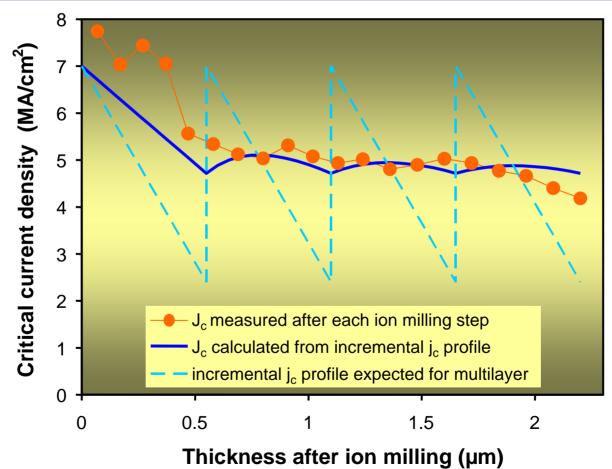


...and this idea suggests that additional interfaces will enable high J_c in thick films...



...by periodically reproducing interfacial enhancement throughout the entire film

Ion milling of multilayer with 4 YBCO layers is consistent with expectations

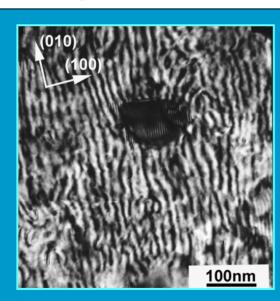


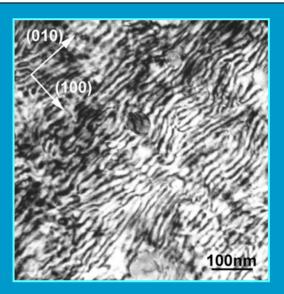


We then began a series of experiments aimed at testing the interfacial enhancement hypothesis

Experiment: Search for something different about the interfacial region. **Result**: Found evidence for misfit dislocations originating at the interface.

TEM plan views of a ~ 20 nm thick YBCO film on a SrTiO₃ single-crystal substrate





Misfit between SrTiO₃ lattice and:

YBCO a-axis - 2.4 %

YBCO b-axis - 0.7 %

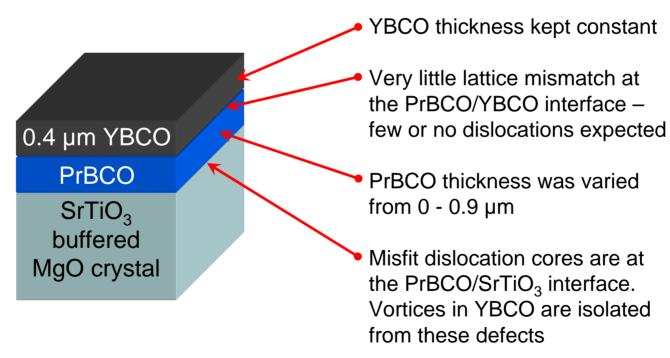
Spacing of ~ 17 nm equals that calculated for a-axis misfit



H. Wang, et al., Physica C, 2006 (in press)

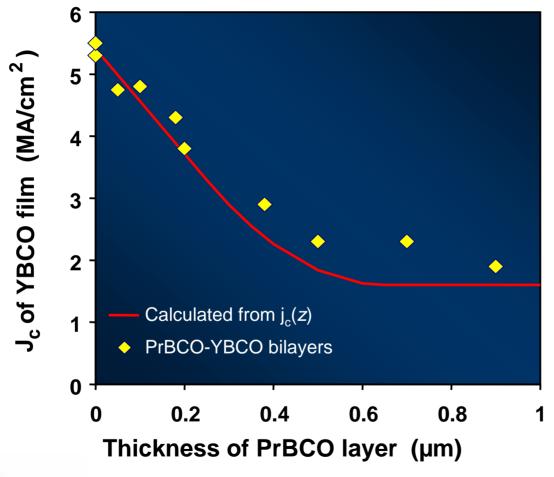
We next measured the effect of separating YBCO from the defect-rich interface...

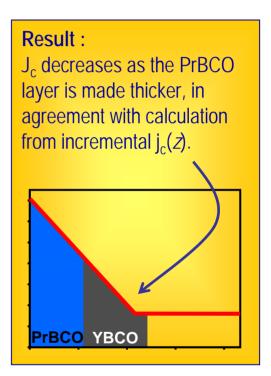
Experiment: Deposit YBCO films of constant thickness on PrBCO layers of different thickness to test the effect of moving the superconductor farther from the substrate interface.



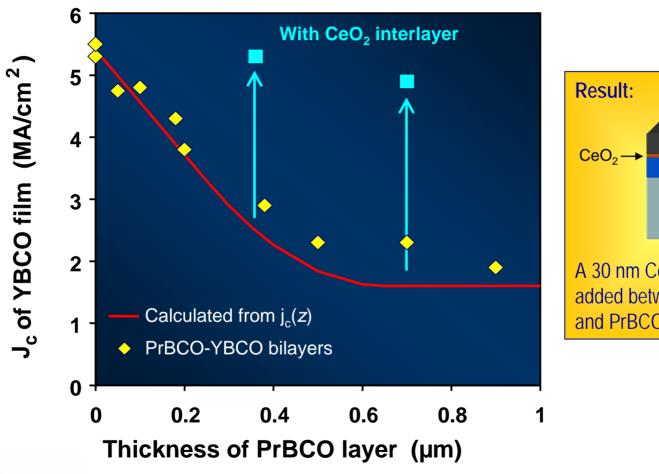


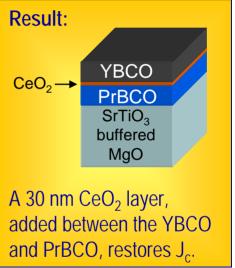
...and found that J_c depends strongly on the distance between the YBCO layer and the defect-rich interface...





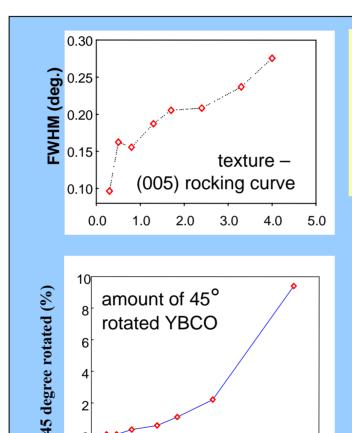
...and that J_c is restored by adding another defect-rich interface under the YBCO





To evaluate the microstructural decay hypothesis we have measured thickness dependence of several properties

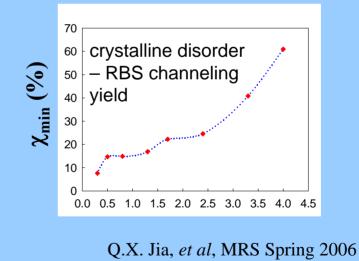
Worse microstructure 🖈



0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5

YBCO on SrTiO₃ substrates

- crystallinity deteriorates as thickness increases
- trend extends over full thickness range
- similar behavior for morphology (roughness, porosity, etc.)

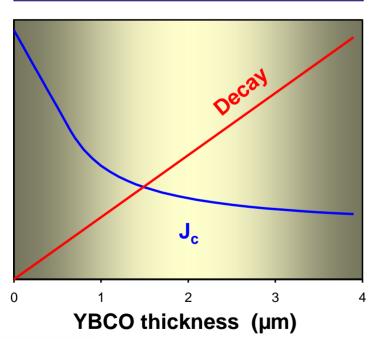




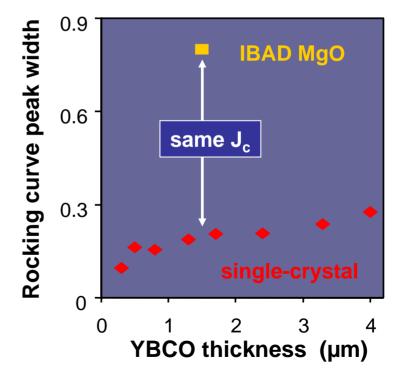
Film thickness (µm)

Assessing the effect of microstructural decay is a difficult business...

★ The observed decay of microstructure does not match the J_c trend for strong thickness dependence.



★ Seemingly important changes in microstructure sometimes produce no change in J_c.





...but even if microstructural decay is not the main factor in strong thickness dependence, it can be a factor in all films

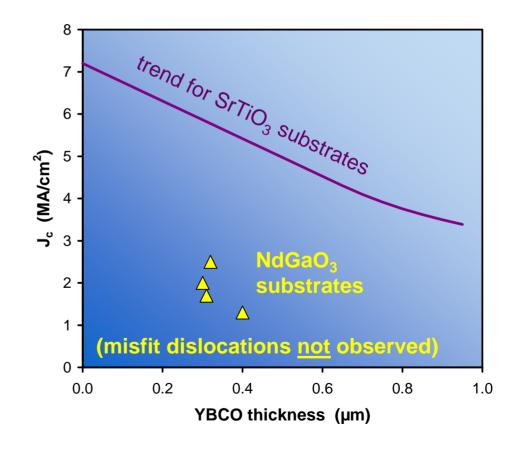
- ★ Because deterioration continues or even accelerates with increasing thickness, microstructural decay must become the dominant current-limiting aspect at some thickness.
- ★ The thickness at which microstructural decay begins to limit J_c is likely dependent on deposition method and processing conditions.



The interfacial enhancement model predicts that J_c will decrease if we eliminate lattice mismatch

Experiment: Investigate the thickness dependence of YBCO on NdGaO₃ substrates.

- (110) NdGaO₃ has an excellent lattice match with YBCO.
- Such a small mismatch can be accommodated without formation of misfit dislocations.
- ♦ Will this affect J_c?





Low J_c and the absence of observable misfit dislocations suggest the following scenario:

- ★ The surface planes of both YBCO and (110) NdGaO₃ have a rectangular lattice.
- **YBCO:** 0.382 nm x 0.388 nm **NdGaO₃:** 0.384 nm x 0.389 nm

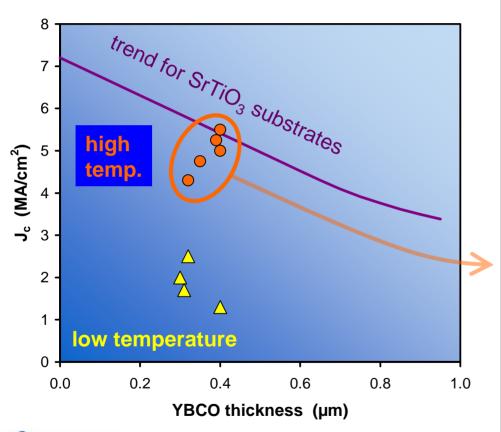
YBCO alignment on NdGaO₃

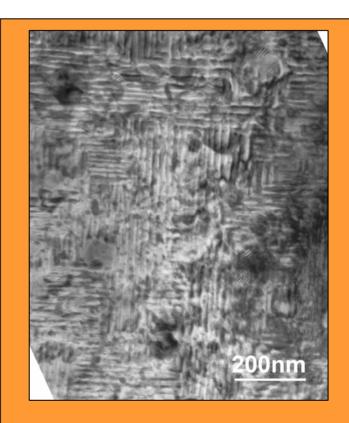
 $a_{YBCO} \parallel a_{NdGaO3} (0.6\%)$ $b_{YBCO} \parallel b_{NdGaO3} (0.15\%)$ small mismatch \rightarrow few dislocations \rightarrow no interfacial enhancement \rightarrow low J_c

But - low J_c can be caused by a number of factors, including the possibility that the deposition temperature is not optimum.



At higher deposition temperature, J_c improved and misfit dislocations were observed at the film-substrate interface

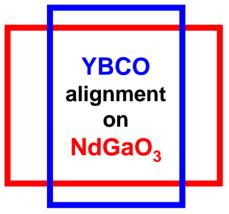




Moiré fringes indicate the presence of misfit dislocations in higher-temperature YBCO films on NdGaO₃



Increased J_c and the presence of misfit dislocations at high temperature suggest the following scenario:



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 $b_{YBCO} || a_{NdGaO3} (1.1\%)$ $a_{YBCO} || b_{NdGaO3} (1.8\%)$ larger mismatch →
high dislocation density →
interfacial enhancement → higher J_c

As unlikely as this epitaxial alignment may seem, there is good evidence that it is occurring in the high-temperature films ->



The dislocation spacing is consistent with the rotated epitaxial relationship

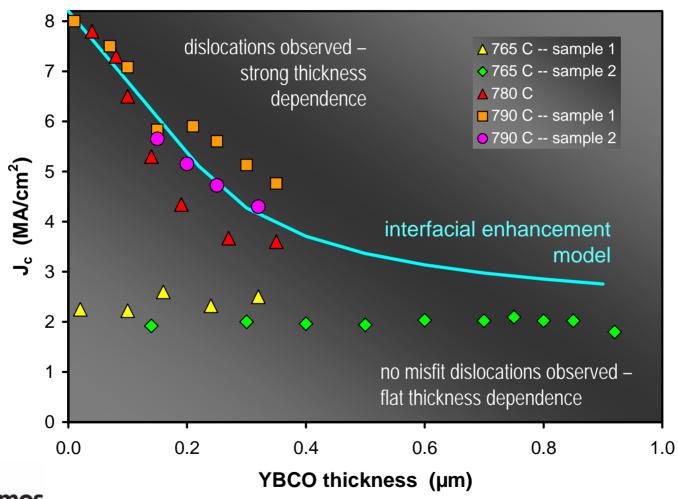
- ★ The predicted dislocation spacing for $a_{YBCO} \parallel b_{NdGaO3}$ is 20.6 nm:
- ★ The observed moiré fringe spacing (same as the misfit dislocation spacing) is ~ 20 nm.
- ★ The amount of mismatch (1.8 %) is comparable to the amount that produces closely-spaced dislocations in:
 - \rightarrow YBCO on SrTiO₃ (2.4 %) or
 - \rightarrow YBCO on CeO₂ (1.7 %).



Moiré fringes in a TEM plan view of a 20 nm-thick YBCO film on NdGaO₃



The main point became clear when ion milled samples of each type showed a correlation between dislocations and J_c(t)



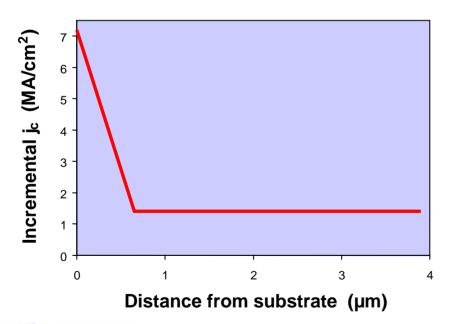
Two significant conclusions emerge from this result

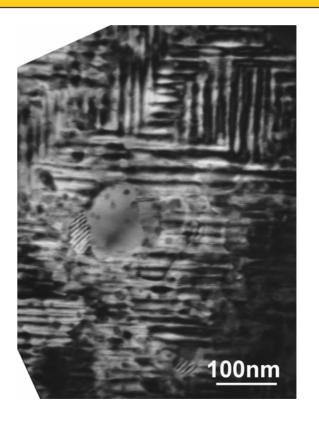
- 1. There is a definite connection between misfit dislocations and strong thickness dependence.
 - ★ However, dislocations may just be an indicator that interfacial enhancement is present, rather than the cause of interfacial enhancement.
- 2. It is possible to find processing conditions that eliminate thickness dependence.
 - This can explain reports in the literature of varying degrees of thickness dependence.
 - ★ This ability will also be a valuable tool in determining the fundamental source of high interfacial J_c.



In summary: There is much evidence in support of the interfacial enhancement model of thickness dependence

- Incremental j_c is constant beyond a certain range and elevated only near the interface.
- 2. The interfacial region has a different microstructure, namely misfit dislocations.







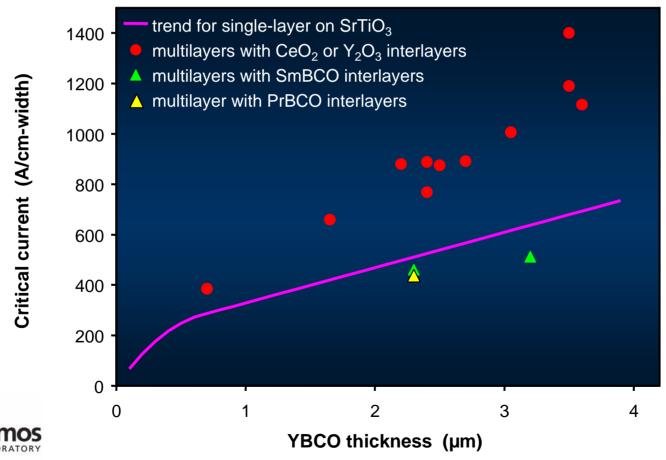
More evidence in support of the interfacial enhancement model of thickness dependence

- 3. Separating YBCO from the defect-rich interface with a PrBCO layer reduces J_c.
- Inserting a CeO₂ layer (and a new defect-rich interface) between YBCO and PrBCO restores J_c.
- 5. Films with misfit dislocations have high interfacial J_c and strong thickness dependence.
- 6. Films with no observable dislocations have flat thickness dependence and no enhancement of J_c near the interface.



More evidence in support of the interfacial enhancement model of thickness dependence

7. Multilayers with heteroepitaxial interfaces "work" – those employing small mismatch have J_c similar to single layer YBCO.



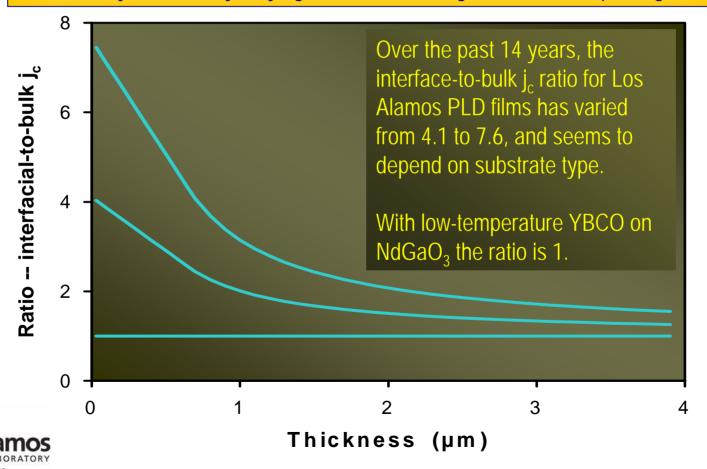
The interfacial enhancement model can explain several other experimental observations

- 1. Different YBCO deposition methods, each producing a unique microstructure, have strikingly similar thickness dependence.
 - * All methods have two aspects in common: a small selection of suitable substrates and roughly the same deposition temperature.
 - ★ These common aspects mean that interfacial stress due to lattice misfit and differential expansion – is approximately the same for all deposition techniques.



The interfacial enhancement model can explain several other experimental observations

 Different degrees of thickness dependence are reported – all can be readily modeled by varying the relative strength of interfacial pinning.



Because of strong evidence for interfacial enhancement our work will now focus on identifying its source

Interfacial enhancement implies a higher degree of beneficial properties near the film-substrate interface. There are several clues that may help identify the desirable properties: For SrTiO₃, YSZ, 1. A confirmed link between MgO, IBAD and misfit dislocations and Incremental j_c (MA/cm²) RABiTS substrates. thickness dependence. $z_r = 0.65 \, \mu \text{m}$ -2. The range of interfacial enhancement is shorter on NdGaO₃ substrates than on others. 0.0 0.5 1.0 2.0 Distance from substrate (µm)



More clues that may help identify desirable interfacial properties

- 3. Higher cation disorder is observed closer to the interface (V. Maroni, this Review).
- 4. Smaller c-axis lattice constant is observed closer to the interface (B. Gibbons, et. al., MRS, Spring 2006).
- 5. Etching reveals higher dislocation density closer to the interface (K. Develos-Bagarinao, *et al.*, Supercond. Sci. Technol. **18**, 667 (2005).

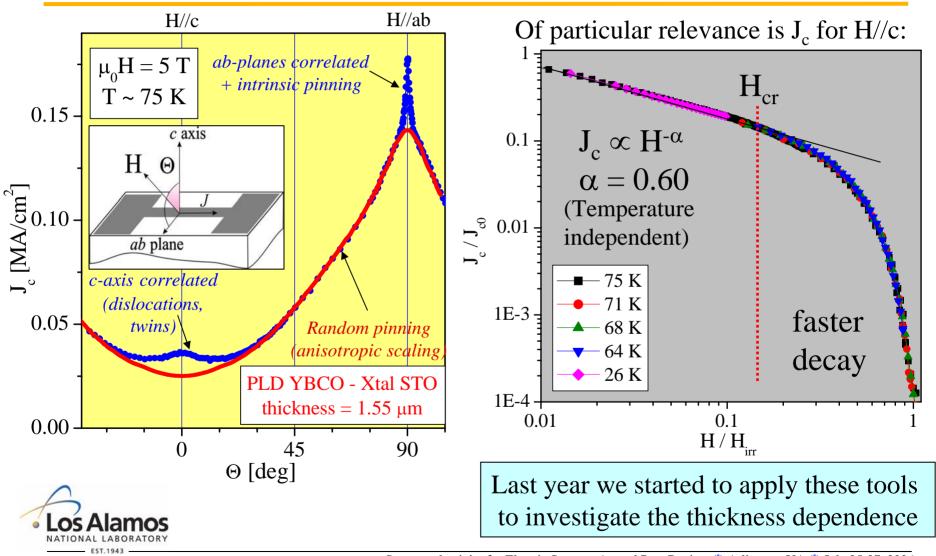


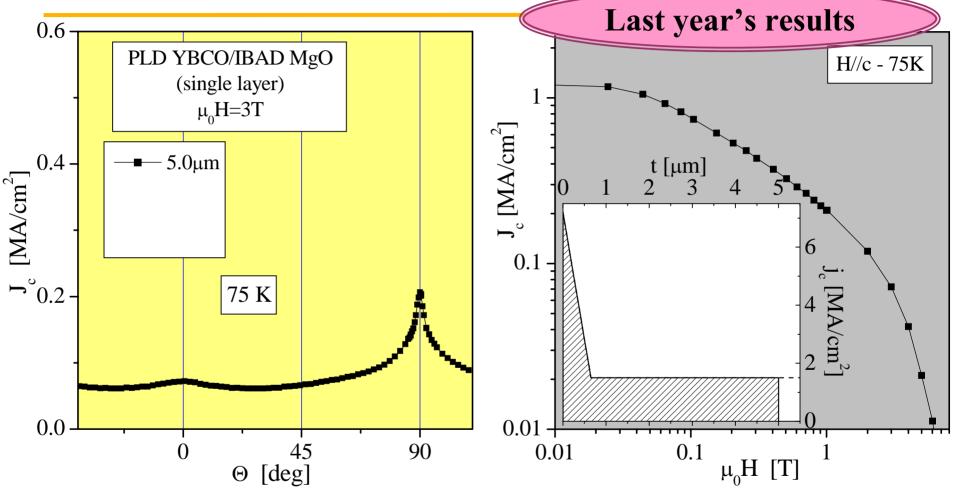
The concept that defects influence the interfacial properties of electronic thin films is widely reported

- ★ The first detailed study took place more than thirty years ago to investigate possible adverse effects on the performance of superlattice devices. ("Defects in epitaxial multilayers, Part 1. Misfit dislocations", J.W. Matthews and A.E. Blakeslee, J. Crystal Growth 27, 118 (1974).)
- ★ There is now a large body of literature reporting interfacial effects on properties of a wide range of electronic film materials.
- ★ The main difference in the case of HTS films is that the interfacial properties are enhanced, not degraded, by defects. But the fact that defects are responsible for improving J_c of YBCO films (e.g., relative to single-crystals) is well-established.

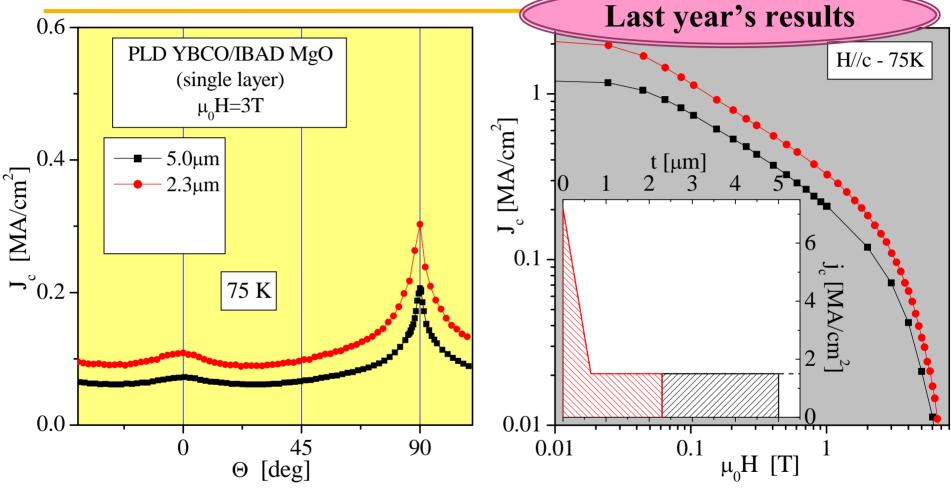


We use the field, angular and temperature dependences of J_c to identify pinning mechanisms and regimes

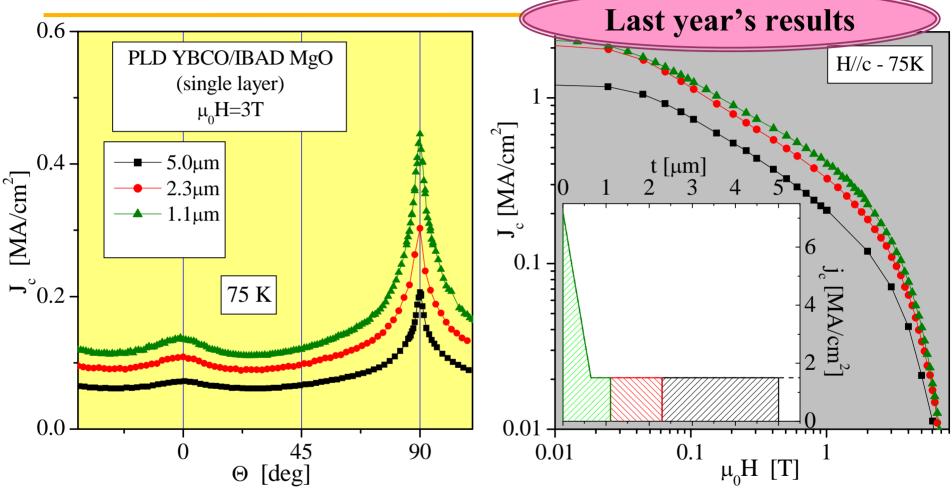




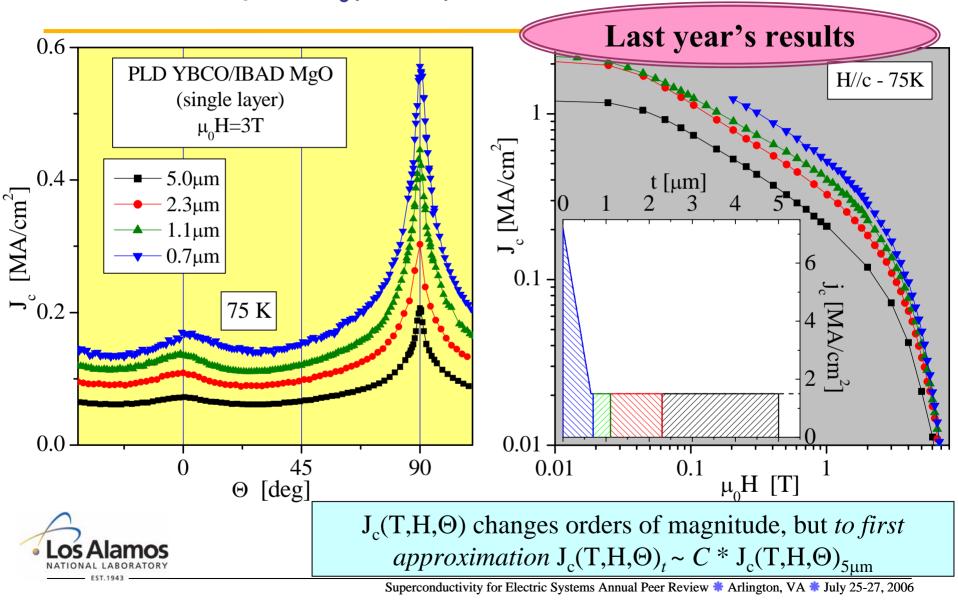




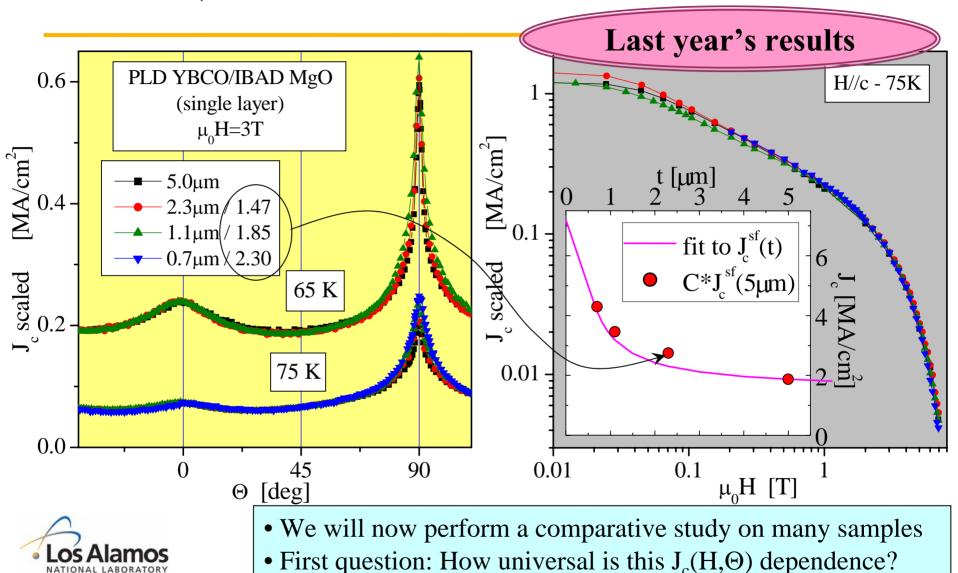




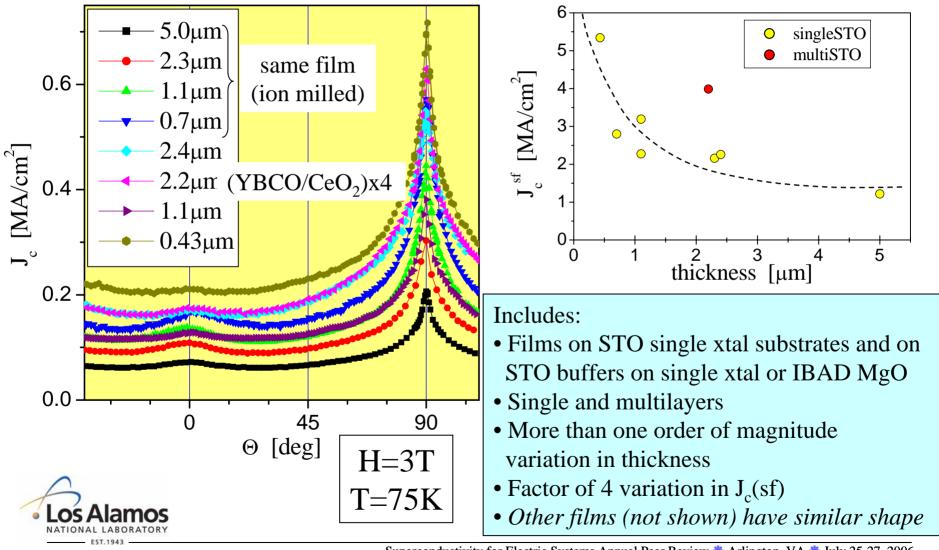




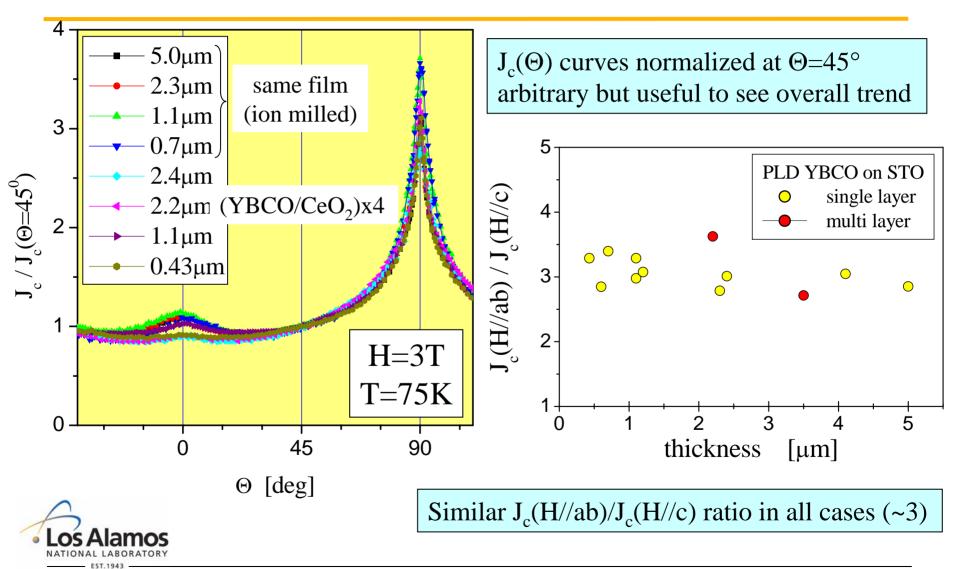
Curves for all thicknesses can be overlapped by just dividing by a factor, which is the same one obtained for self field



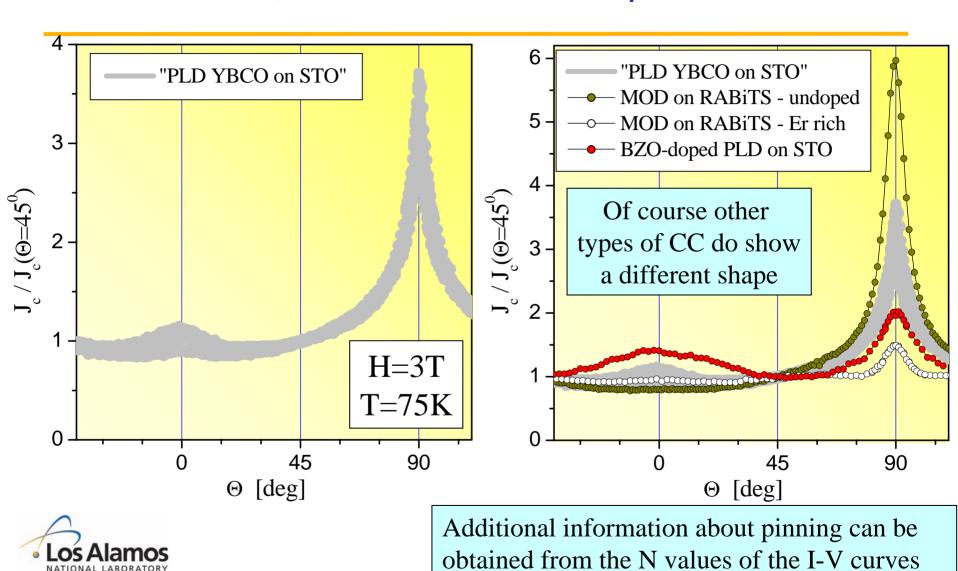
The *shape* of $J_c(\Theta)$ is similar for a wide range of PLD YBCO films on STO



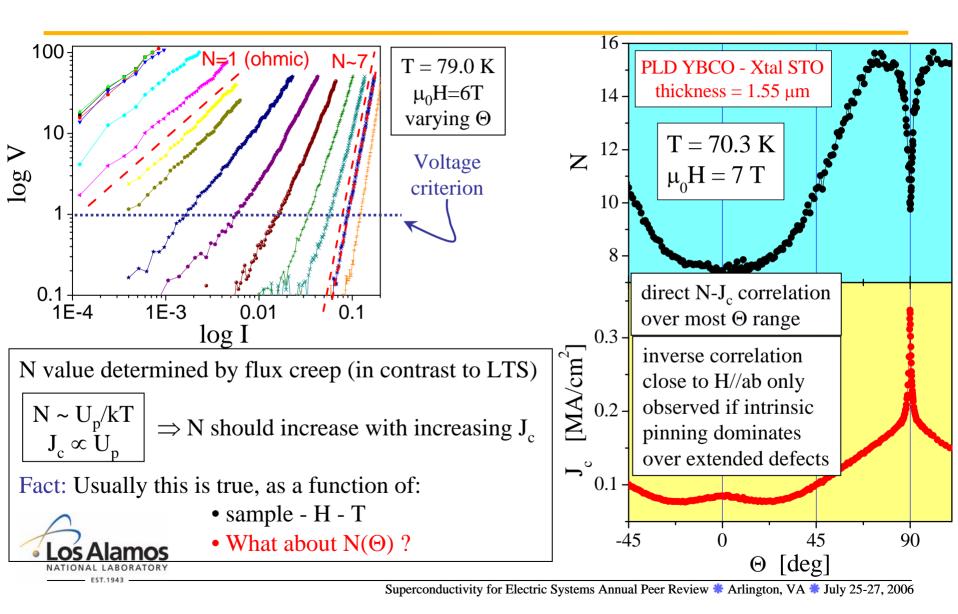
Normalized $J_c(\Theta)$ curves almost overlap: Similar anisotropic random pinning. Some variations in peaks for H//c and H//ab



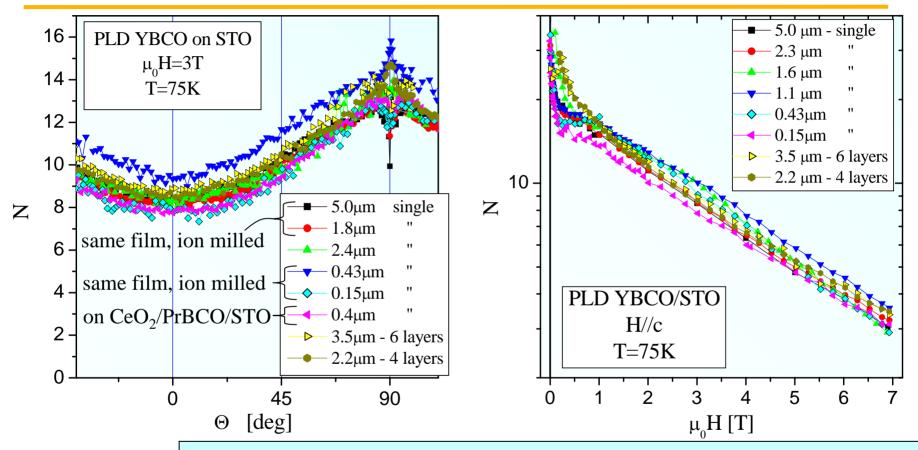
We can build a fuzzy "normalized $J_c(\Theta)$ curve" for PLD YBCO on STO, to be used for later comparisons



I-V curves are well described by a power law V∞I^N What can we learn from N(T,H,Θ)?



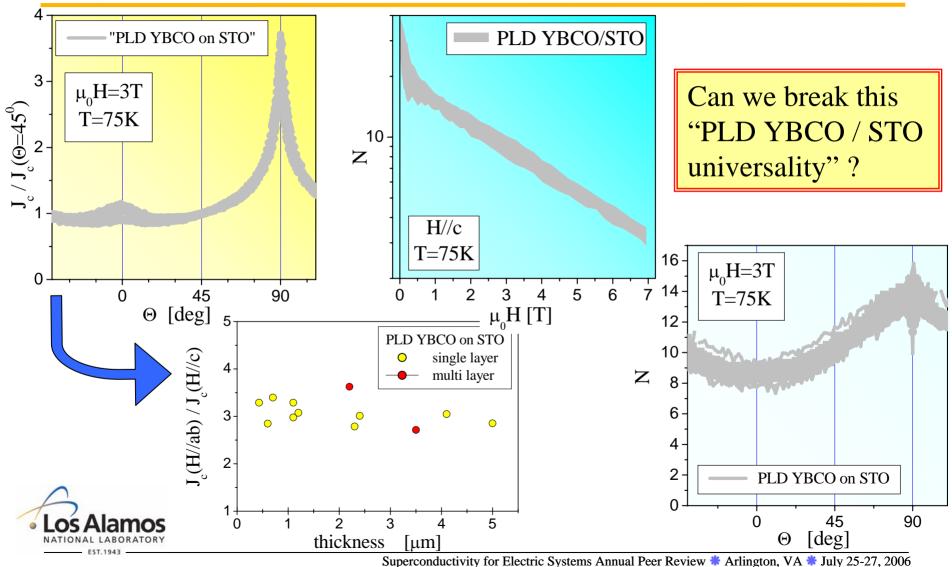
The N(H, Θ) values are almost the same for all samples: Similar activation energy U_p(H, Θ) in all cases



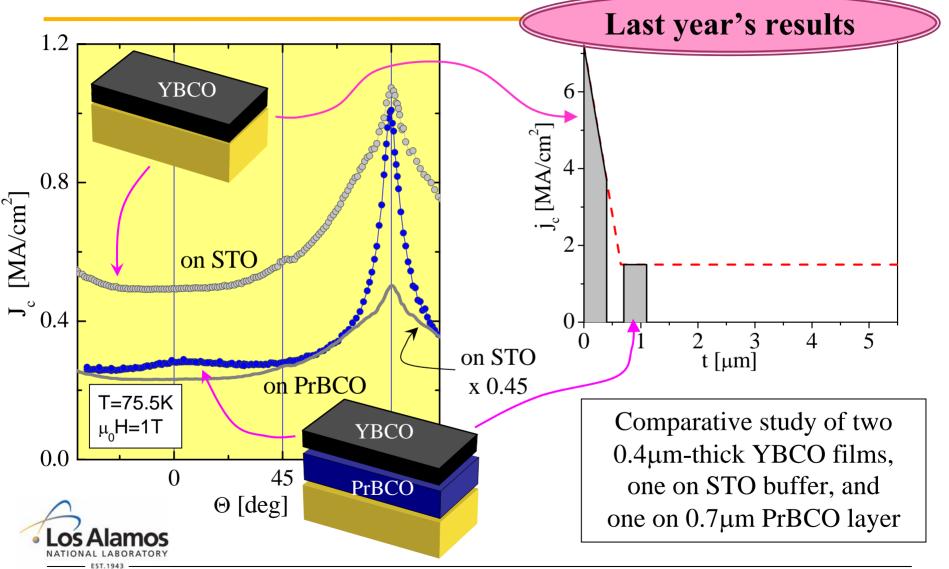
The largest variability is observed:

- At low fields
- Near H//ab (intrinsic pinning vs extended defects, shown in PR05)

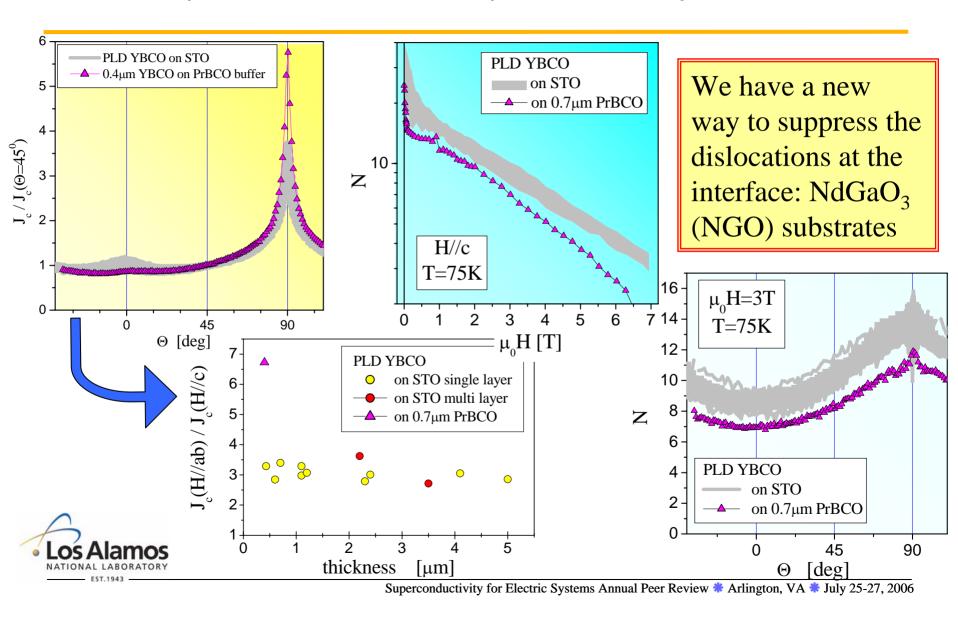
Summary: evidence indicates that the dominant pinning mechanisms are the same for all PLD YBCO/STO films (thickness from 0.15 to 5 mm, single- and multi-layers)



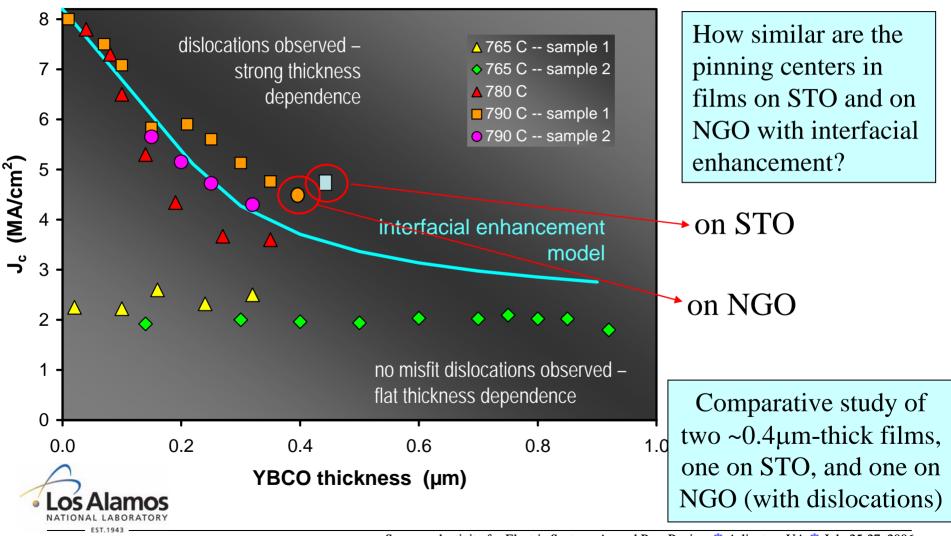
A PrBCO spacer layer separating the YBCO from the STO interface eliminates the J_c enhancement



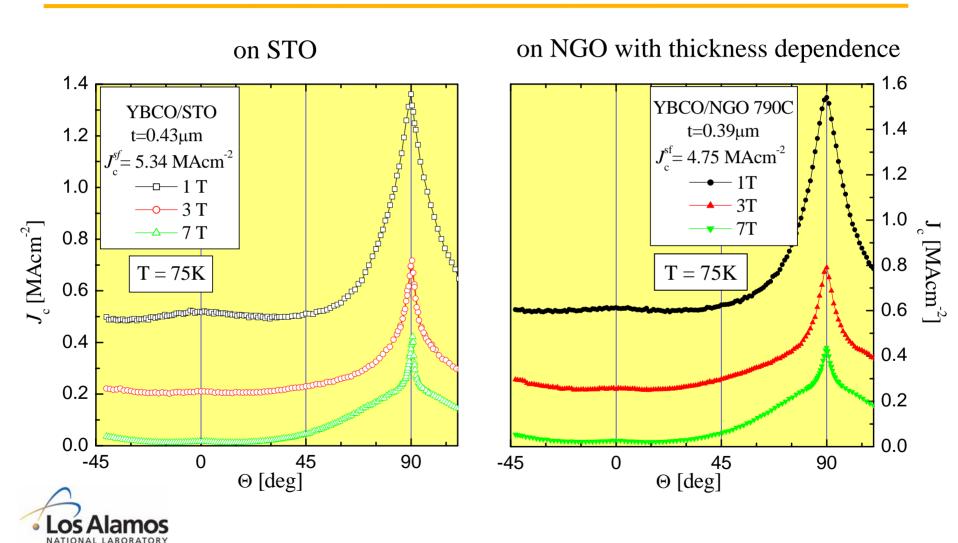
The angular and field dependences of J_c and N of the $0.4\mu m$ thick film on a $0.7\mu m$ PrBCO layer *are* different



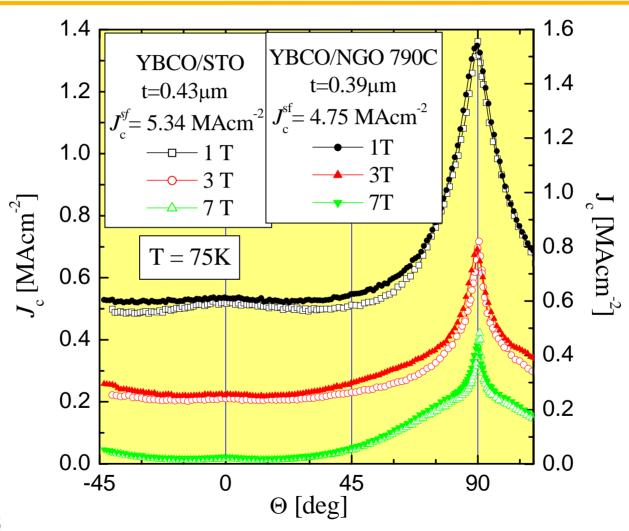
The NdGaO₃ substrates open the opportunity to compare films with and without misfit dislocations at the interface



"Validation" of the NGO: $J_c(H,\Theta)$ in both ~0.4 μ m films (on STO and on NGO with interface dislocations) are the same

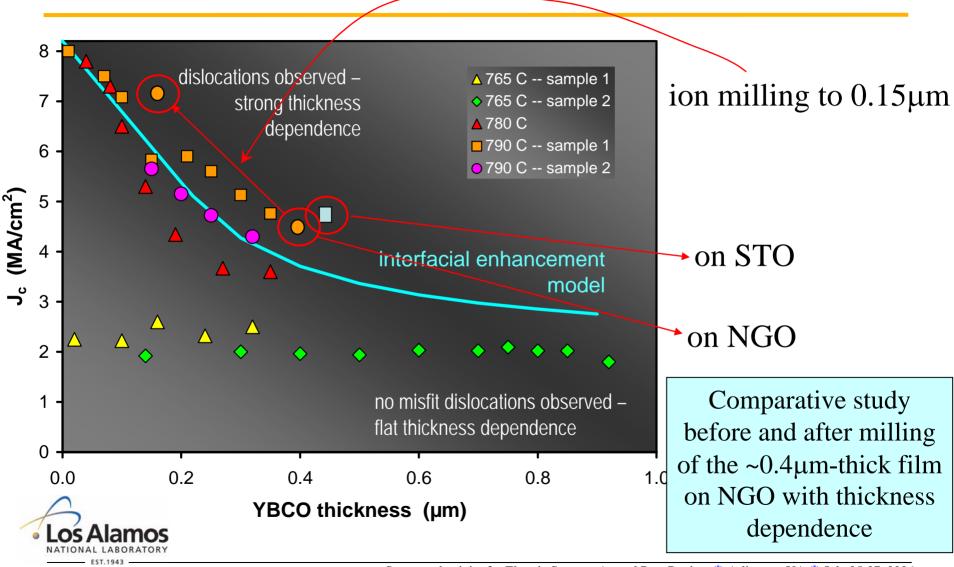


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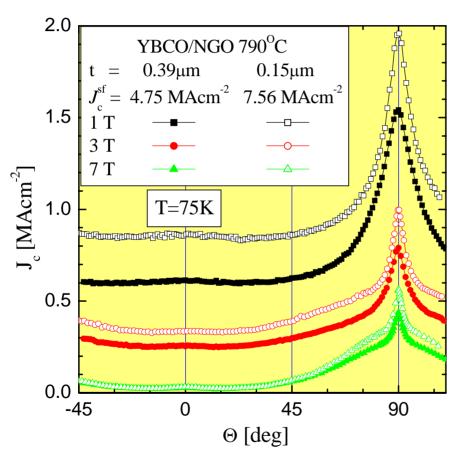


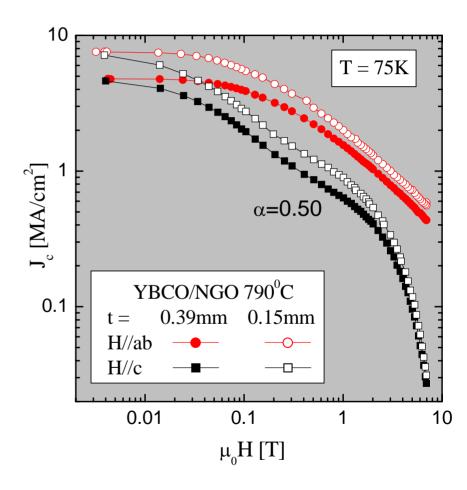


Does the universal behavior extend to thinner films on NdGaO₃ substrates ?



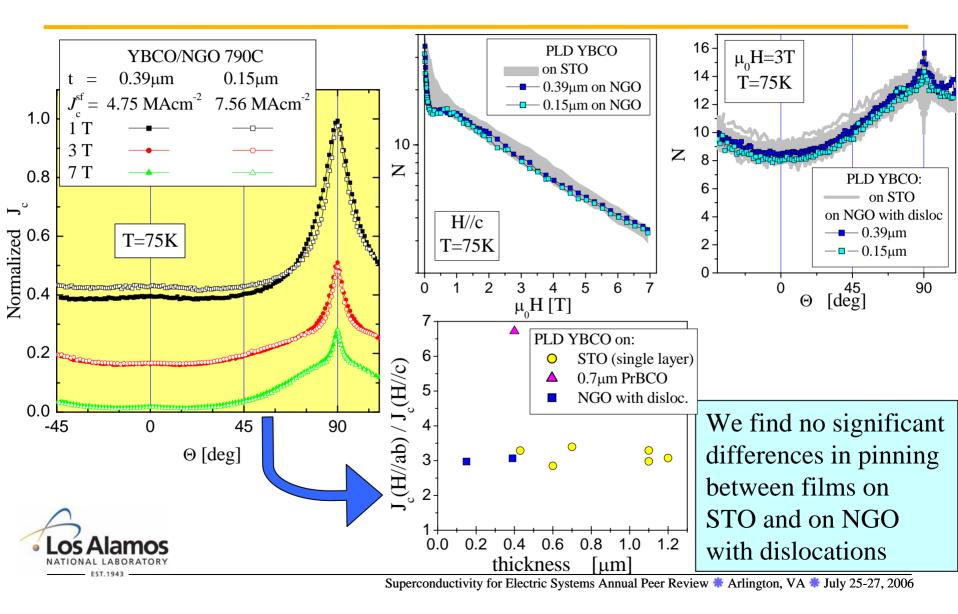
The NGO film ion-milled to 0.15 μm still retains the same angular and field dependence of J_c and N



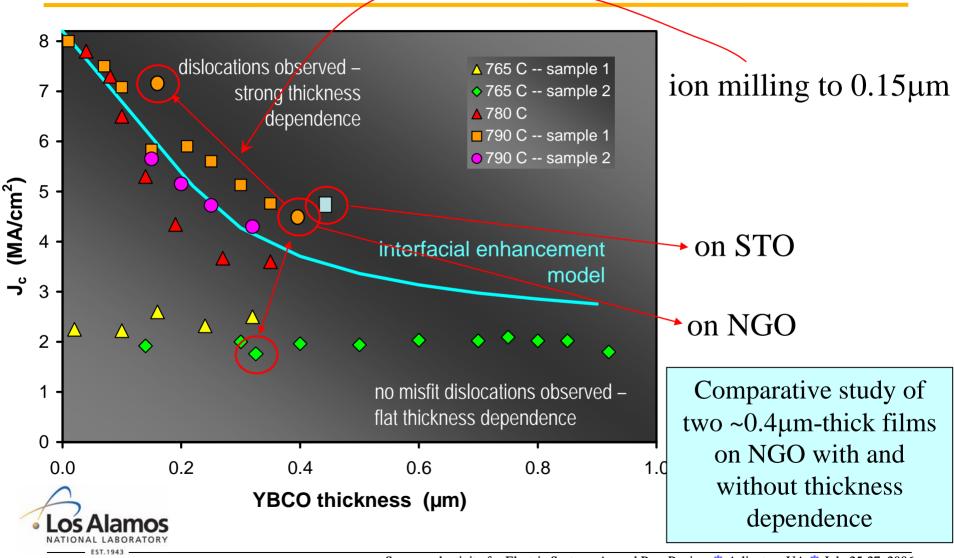




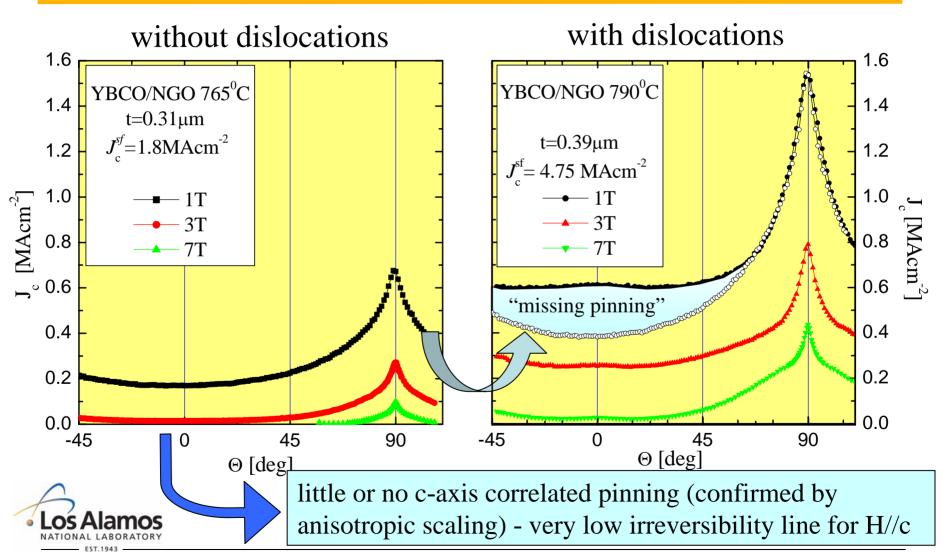
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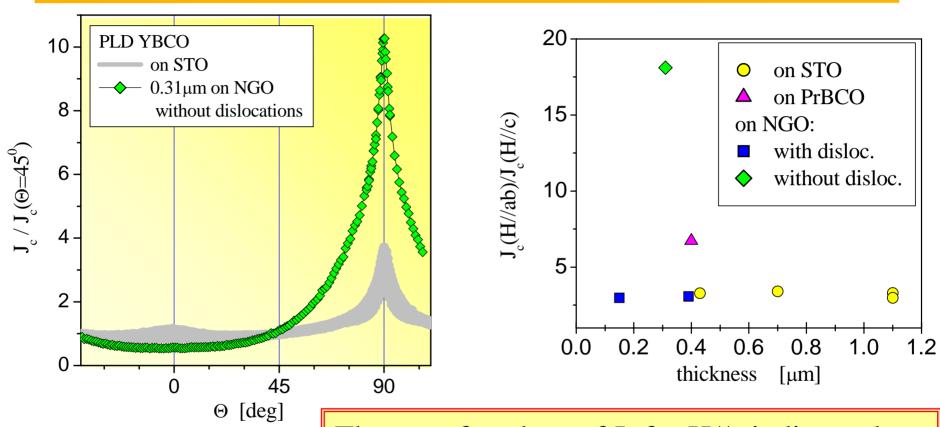
The NdGaO₃ substrates open the opportunity to compare films with and without misfit dislocations at the interface



The film on NGO without interface dislocations not only has lower $J_c(sf)$, but also a very different shape of $J_c(\Theta)$



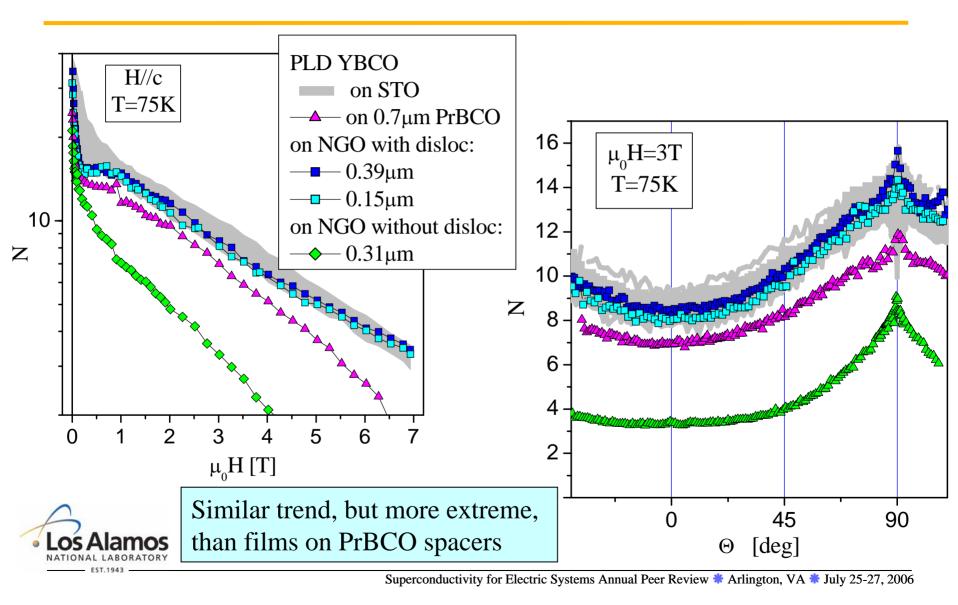
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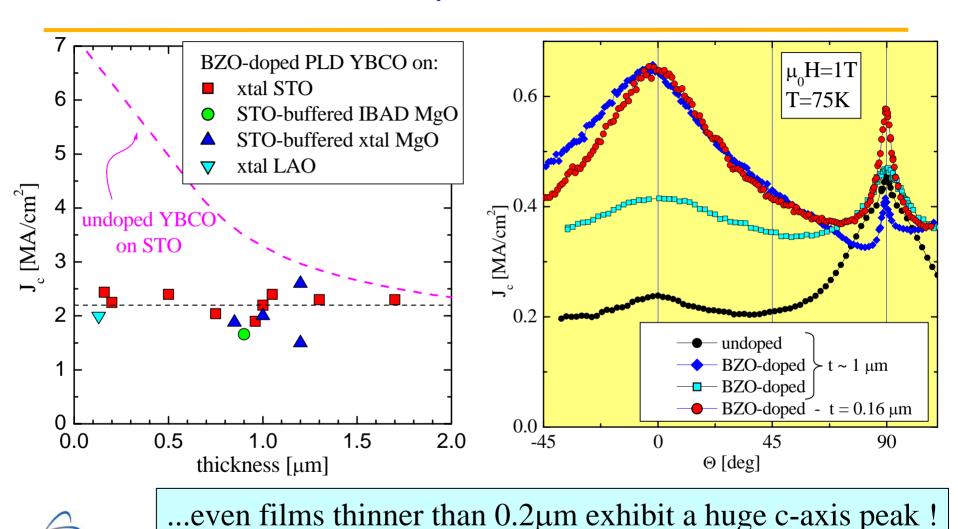


The very fast drop of J_c for H//c indicates low pinning by random defects, which have lower irreversibility line than correlated defects.

The much lower N values in the NGO film without misfit dislocations also confirms the low pinning energy

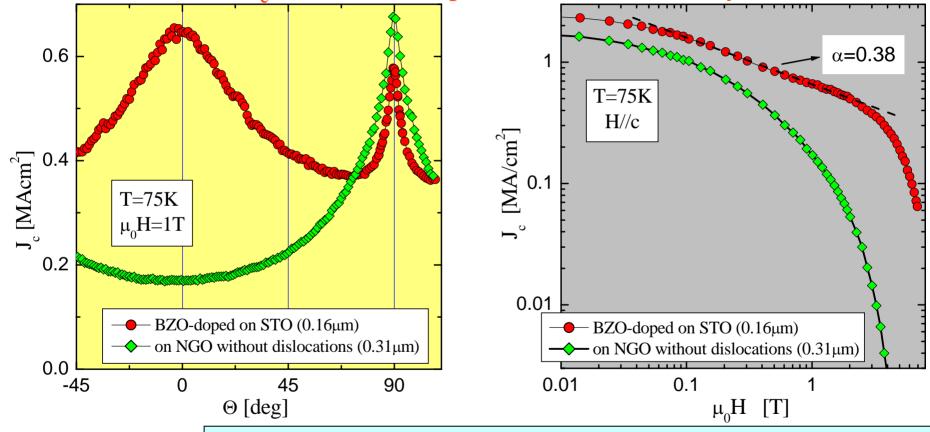


The J_c(sf) of our BaZrO₃ (BZO) doped films has no thickness dependence, however...



BZO-doped films and (undoped) films on NGO without interface dislocations have in common the absence of thickness dependence and similar $J_c(sf)$ values, but...

...the in-field J_c of the BZO-doped films is dramatically better



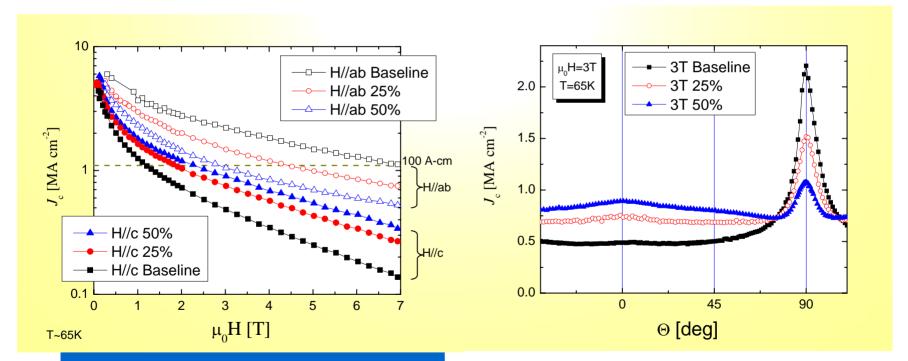


Some mechanism suppresses the interface enhancement in the BZO-doped films, but the strong c-axis correlated pinning is still present

Substantial improvements for $J_c(H//c)$ were found for Er_2O_3 additions up to the level of 50% $(Y_1Er_{0.5}Ba_2Cu_3O_v)$.

- Little effect on J_c(self-field)
- Overall increase in J_c(H//c) at the expense of J_c(H//ab)
- What are the changes in the pinning microstructure?

```
Baseline (Y_1Ba_2Cu_3O_y) = \frac{I_c(sf,75K)}{25\%Er} (Y_1Er_{0.25}Ba_2Cu_3O_y) = 260 A/cm 50\%Er (Y_1Er_{0.5}Ba_2Cu_3O_y) = 260 A/cm
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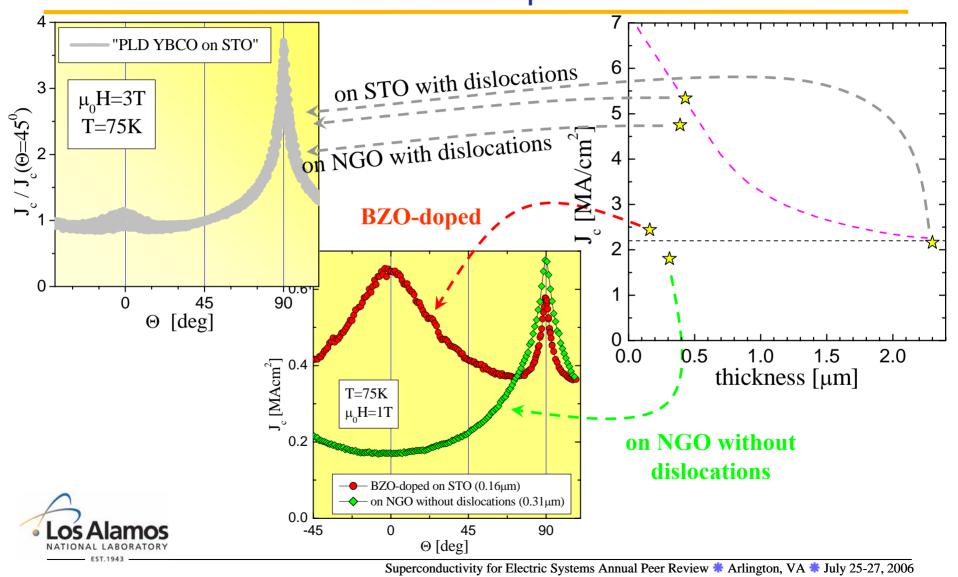
Same trends in $J_c(H,\Theta)$ at 65K and 75K

Leonardo Civale and Boris Maiorov (LANL)



presented by T. Holesinger in the WDG talk at PR05

Summary: We find universal $J_c(H,\Theta)$ behavior in PLD films with interface enhancement, but $J_c(H,\Theta)$ in films without enhancement is not unique



Scoring Criterion - Results

- 1. Demonstrated that interfacial enhancement is responsible for strong thickness dependence and is consistent with most experimental observations of thickness dependence.
- 2. Established a definite link between interfacial misfit dislocations and thickness dependence.
- 3. Found two processing conditions for which thickness dependence is eliminated (low temperature YBCO on NdGaO₃ and BaZrO₃-doped YBCO).
- 4. Established universal behavior of $J_c(H,\Theta)$ for PLD YBCO single- and multi-layers on SrTiO₃ (xtal or buffer), over a wide thickness range.



Scoring Criterion - Results (continued)

- 5. Studied H,⊕ dependence of exponent N of power law I-V curves (associated with activation energy) also found universal behavior.
- 6. Showed that $J_c(H,\Theta)$ and $N(H,\Theta)$ for films on $NdGaO_3$ with thickness dependence follow the same behavior as films on $SrTiO_3$.
- 7. Determined that films on $NdGaO_3$ and PrBCO without thickness dependence show different $Jc(H,\Theta)$ and $N(H,\Theta)$ behavior: faster decay with field, less pinning for field orientations near c-axis.
- 8. Moved to higher-J_c multilayers, achieving 990 A/cm-width in a 2.2 µm thick film.



Scoring criterion - Research Integration

- ★ In 2006 we worked with SuperPower to demonstrate the compatibility of high-current multilayer technology with the MOCVD process, and engaged in characterization and analysis of flux pinning in MOCVD films (CRADA).
- ★ Continued our participation in the Wire Development Group. We performed extensive structural and electrical characterization, and contributed to advancing the understanding of pinning enhancement methods. Developed new capabilities for long-length characterization (presented in Wire Session).
- We established a CRADA with AMBP Technologies to study a novel method for adding flux-pinning defects to YBCO.



Scoring criterion - Research Integration (continued)

- ★ We collaborated with Argonne National Laboratory in the analysis of thickness dependence of cation disorder.
- ★ We continued our long-standing collaboration with University of Cambridge in the area of flux-pinning enhancement and in the study of Hybrid Liquid Phase Epitaxy as an alternate YBCO deposition method.
- ★ We continued our collaboration with the Applied Superconductivity Center at University of Wisconsin, Madison on flux-pinning studies (mutual visits, sample exchanges and joint presentations).
- ★ We began a collaboration with the National High Magnetic Field Laboratory/Florida State University to study high-field vortex pinning and performance of coated conductors.



Scoring Criterion - Performance

Plan: Continue research into the cause of elevated j_c near the interface.

Goal: Understand the 0.65 µm range of influence.

Accomplishments:

- Established conclusively that thickness dependence arises from interfacial enhancement.
- Showed that interfacial enhancement and misfit dislocations are strongly correlated
- Found a way to change the 0.65 µm range of influence...or eliminate it altogether.
- Used angular and field dependence to demonstrate universal behavior for PLD films with strong thickness dependence from 0.15 – 5 µm thickness.
- Showed a different behavior for films on NdGaO₃ and PrBCO that have no thickness dependence.



Plan: Survey alternate interlayer or buffer layer materials.

Goal: Determine which properties are significant in producing high interfacial j_c.

Accomplishments:

- Determined that lattice mismatch is an important property by showing that multilayers using CeO₂ or Y₂O₃ interlayers have significantly elevated J_c, while ones using small-mismatch interlayers (SmBCO, PrBCO) do not.
- Demonstrated strong interfacial enhancement occurs when the mismatch between buffer layer and *either* YBCO axis is 1.7 – 2.4 %.
- Proved that interfacial enhancement (and thickness dependence) can be eliminated if lattice mismatch is sufficiently reduced.



Plan: Push the practical limit to thick-film I_c that can be achieved by depositing a greater number of thinner YBCO layers.

Goal: 1000 A/cm-width in a 2 µm film.

Accomplishment:

990 A/cm-width at 2.2 μm.



Plan: Combine multilayers with an in-field pinning enhancement method.

Goal: self-field $I_c = 1000$ A/cm-width and $\alpha < 0.4$.

Accomplishments:

- Made multilayers using BaZrO₃-doped YBCO and found that there was no improvement over a single-layer of the same total thickness.
- To diagnose, checked the thickness dependence of BaZrO₃-doped YBCO and found that it is very weak, or absent.
- Hypothesized that weak thickness dependence arises because the BaZrO₃
 particles provide an alternate means to relieve interfacial stress, or otherwise
 interrupt the formation of misfit dislocations.



Plan: Work closely with SuperPower to produce high-current multilayers on IBAD MgO using MOCVD.

Goal: Significant improvement over single-layer I_cs.

Accomplishment:

- Demonstrated the feasibility of using the multilayer approach with MOCVD, especially in concert with SuperPower's multipass method. Incremental improvement was realized, but was overshadowed by substantial progress in single-layer J_c at SuperPower.
- This effort will continue in FY 2007.



Plan: Continue to work with American Superconductor in the understanding and enhancement of vortex pinning in ex-situ films.

Goal: To be coordinated with AMSC.

Accomplishment:

 Our work in this area was described in the Wire Development Group presentation to the Joint Panel, and in the presentation by Holesinger and Civale in the Wire Session



Scoring Criterion - FY 2007 Plans

Pursue the link between interfacial defects and thickness dependence to determine the source of interfacial enhancement of J_c:

- → Establish a collaboration to theoretically examine the film stresses leading to formation of misfit dislocations, and the nature of the dislocations themselves.
- → Design experiments to systematically change film stresses and measure the effect on thickness dependence.
- → Conduct microscopic examinations of films to illuminate the nature and extent of defects present in the interfacial region.
- → In combination with microscopic examination, use field and angular dependence to identify the defects responsible for both thickness dependence and in-field pinning improvement.



Scoring Criterion - FY 2007 Plans (continued)

- ★ Find the reason for flat thickness dependence in BaZrO₃-doped YBCO films.
- ★ Use the above result to find an in-field pinning enhancement method compatible with high-current multilayers.

